

1. A method of preparing a pyrrolotriazin-4-one compound represented by the following general formula (4), the method comprising:

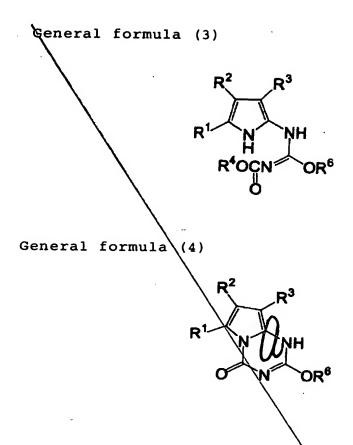
an addition step of reacting an aminopyrrole derivative represented by the following general formula (1) with a reactant represented by the following general formula (2) for forming an adduct represented by the following general formula (3); and

a cyclization step of cyclizing the adduct represented by the following general formula (3) for forming the pyrrolotriazin-4-one represented by the following general formula (4),

General formula (1)

$$R^2$$
 R^3
 N
 N
 N

General formula (2)



wherein, in the general formulas, R¹ represents a hydrogen atom, an alkyl group, an aryl group, or a group capable of withdrawing; R² and R³ each independently represents a hydrogen atom, an alkyl group, an aryl group, a cyano group, a substituted sulfonyl group, a substituted carbonyl group, or a halogen atom; R⁴ represents a substituted or non-substituted alkyl group, or a substituted or non-substituted aryl group; R⁵ represents a substituted or non-substituted alkyl group, a substituted or non-substituted alkyl group, a substituted or non-substituted aryl group, or a substituted or non-substituted heterocyclic group; and R⁶ represents a substituted or non-substituted alkyl group having at least 3 carbon atoms, or a substituted or non-substituted or non-substituted aryl group.

2. The method of preparing a pyrrolotriazin-4-one

according to claim 1, wherein at least one of a first acid, a salt of the first acid, and a base is present in a reaction system of the addition step.

- The method of preparing a pyrrolotriazin-4-one according to claim 2, wherein the first acid is at least one acid selected from the group consisting of alkylsulfonic acid, arylsulfonic acid, alkylcarboxylic acid and Lewis acid.
- 4. The method of preparing a pyrrolotriazin-4-one according to claim 2, wherein the salt of the first acid is at least one salt of the acid selected from the group consisting of alkylsulfonic acid, arylsulfonic acid, alkylcarboxylic acid and Lewis acid.
- 5. The method of preparing a pyrrolotriazin-4-one according to claim 1, wherein at least one of a second acid, a salt of the second acid, and a base is present in a reaction system of the cyclization step.
- 6. The method of preparing a pyrrolotriazin-4-one according to claim 2, wherein at least one of a second acid, a salt of the second acid, and a base is present in a reaction system of the cyclization step.
- 7. The method of preparing a pyrrolotriazin-4-one according to claim 3, wherein at least one of a second acid, a salt of the second acid, and a base is present in a reaction system of the cyclization step.
- 8. An isothiocyanatoformic acid ester derivative represented by the following general formula (2)

Seneral formula (2)

in which: R' represents a substituted or non-substituted alkyl group, or a substituted or non-substituted aryl group; R⁵ represents a substituted or non-substituted alkyl group, a substituted or non-substituted aryl group, or a substituted or non-substituted ferocyclic group; and R⁶ represents a substituted or non-substituted alkyl group having at least 3 carbon atoms, or a substituted or non-substituted aryl group

A method of preparing the isothiocyanatoformic acid ester derivative according to claim 8, the method comprising the steps of:

adding a chloroformic acid derivative represented by the following general formula (7) to an isothiocyanic acid salt represented by the following general formula (5) and a hydroxy derivative represented by the following general formula (6) for preparing an intermediate represented by the following general formula (8); and

preparing the isothiocyanatoformic acid ester derivative from the intermediate:

ZNCS

General formula (5)

R⁶OH

General formula (8

contd Q²

CICQR⁴

O S R⁴OCNHCOR⁶

General formula (7)

General formula (8)

wherein, in the general formula (5), Z represents a sodium atom or a potassium atom

10. A method of preparing the isothiocyanatoformic acid ester derivative according to claim g, the method comprising the steps of:

preparing a first intermediate represented by the following general formula (8);

preparing a second intermediate represented by the following general formula (10) from the first intermediate and a compound represented by the following general formula (9); and

preparing the isothiocyanatoformic acid ester derivative from the second intermediate:

O S R⁴OCNHCOR⁶

M(OH)

General formula (8)

General formula (9

contd a²

General formula (10)

wherein, in the general formulas (9) and (10), M represents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom.

11. A method of preparing the isothiocyanatoformic acid ester derivative according to claim 8, the method comprising the steps of:

preparing an intermediate represented by the following general formula (10); and

reacting the intermediate with an alkylating agent represented by one of the following general formula (11) and the following general formula (12) for preparing the isothiocyanatoformic acid ester derivative:

$$\begin{pmatrix}
 O & OR^6 \\
 R^4OCN = C & S & M$$

General formula (10)

R⁵X

General formula (11)

 $(R^{5}O)_{2}SO_{2}$

General formula (12)

contol a^2

wherein, in the general formula (10), M represents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom, and, in the general formula (11), X represents a halogen atom or SO₃Ar, and Ar represents a substituted or non-substituted aryl group.

12. A method of preparing the isothiocyanatoformic acid ester derivative according to claim 8, the method comprising the steps of:

adding a chloroformic acid derivative represented by the following general formula (7) to an isothiocyanic acid salt represented by the following general formula (5) and a hydroxy derivative represented by the following general formula (6) for preparing a first intermediate represented by the following general formula (8);

preparing a second intermediate represented by the following general formula (10) from the first intermediate and a compound represented by the following general formula (9); and

preparing the isothiocyanatoformic acid ester derivative from the second intermediate:

ZNCS

 R^6OH

General formula (5)

General formula (6)

contd.

CICOR4

General formula (7)

O S R⁴OCNHCOR⁶

General formula (8)

 $M(OH)_n$

General formula (9)

R4OCN=C S N M

General formula (10)

wherein, in the general formula (5), Z represents a sodium atom or a potassium atom, and, in the general formulas (9) and (10), M represents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom.

13. A method of preparing the isothiocyanatoformic acid ester derivative according to claim 8, the method comprising the steps of:

preparing a first intermediate represented by the following general formula (8);

preparing a second intermediate represented by the following general formula (10) from the first intermediate and a compound represented by the following general formula (9); and

reacting the second intermediate with an alkylating agent represented by one of the following general formula (11) and the following general formula (12) for preparing the isothiocyanatoformic acid ester derivative:

contd a²

O S R⁴OCNHCOR⁶

General formula (8)

 $M(OH)_n$

General formula (9)

R4OCN=C S N M

General formula (10)

R⁵X

General formula (11)

(R⁵O)₂SO₂

General formula (12)

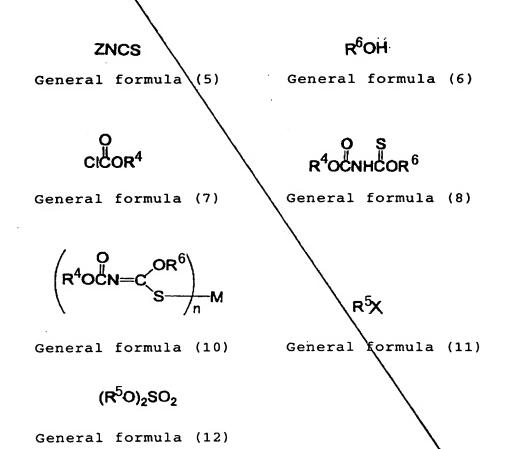
wherein, in the general formulas (9) and (10), Mrepresents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom, and, in the general formula (11), X represents a halogen atom or SO₃Ar, and Ar represents a substituted or non-substituted aryl group

14. A method of preparing the isothiocyanatoformic acid ester derivative according to claim 8, the method comprising the steps of:

adding a chloroformic acid derivative represented by the following general formula (7) to an isothiocyanic acid salt represented by the following general formula (5) and a hydroxy derivative represented by the following general formula (6) for preparing a first intermediate represented by the following general formula (8);

contol.

preparing a second intermediate represented by the following general formula (10) from the first intermediate; and reacting the second intermediate with an alkylating agent represented by one of the following general formula (11) and the following general formula (12) for preparing the isothiocyanatoformic acid ester derivative:



wherein, in the general formula (5), Z represents a sodium atom or a potassium atom, in the general formula (10), M represents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom, and, in the general formula

contd.

(11), X represents a halogen atom or SO₃Ar, and Ar represents a substituted or non-substituted aryl group.

15. A method of preparing the isothiocyanatoformic acid ester derivative according to claim 8, the method comprising the steps of:

adding a chloroformic acid derivative represented by the following general formula (7) to an isothiocyanic acid salt represented by the following general formula (5) and a hydroxy derivative represented by the following general formula (6) for preparing a first intermediate represented by the following general formula (8);

preparing a second intermediate represented by the following general formula (10) from the first intermediate and a compound represented by the following general formula (9); and

reacting the second intermediate with an alkylating agent represented by one of the following general formula (11) and the following general formula (12) for preparing the isothiocyanatoformic acid ester derivative:

ZNCS

R⁶OH

General formula (5)

General formula (6)

CICOR⁴

General formula (7)

General formula (8)

contd a

 $M(OH)_n$

General formula (9)

Q OR6 OR6 N M

General formula (10)

· R⁵X

(R⁵O)₂SO₂

General formula (11)

General formula (12)

wherein, in the general formula (5), Z represents a sodium atom or a potassium atom, in the general formulas (9) and (10), M represents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom, and, in the general formula (11) X represents a halogen atom or SO₃Ar, and Ar represents a substituted or non-substituted aryl group.